LLNL-JRNL-540571

LAWRENCE LIVERMORE NATIONAL LABORATORY

SNM Detection with an Optimized Water Cherenkov Neutron Detector

S. Dazeley, A. Bernstein, M. Sweany

March 22, 2012

Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment

SNM Detection with an Optimized Water Cherenkov Neutron Detector

S. Dazeley*, A. Bernstein, M. Sweany
Lawrence Livermore National Laboratory, Livermore, CA 94550, USA

Abstract:
Special Nuclear Material (SNM) can either spontaneously fission or be induced to do so, either case results in neutron emission. For this reason, neutron detection performs a crucial role in the functionality of Radiation Portal Monitoring (RPM) devices. Since neutrons are highly penetrating and difficult to shield, they could potentially be detected escaping even a well-shielded cargo container. If the shielding were sophisticated, detecting escaping neutrons would require a highly efficient detector with close to full solid angle coverage. In 2008 we reported the successful detection of neutrons with a ¼ tonne gadolinium doped water Cherenkov prototype [1] – a technology that could potentially be employed cost effectively with full solid angle coverage. More recently we have built and tested both 1-kiloliter and 3.5-kiloliter versions [2], demonstrating that very large, cost effective, non-flammable and environmentally benign neutron detectors can be operated efficiently without being overwhelmed by background. In this paper we present a new design for a modular system of water based neutron detectors that could be deployed as a real RPM. The modules contain a number of optimizations that have not previously been combined within a single system. We present simulations of the new system, based on the performance of our previous detectors. Our simulations indicate that an optimized system such as is presented here could achieve SNM sensitivity competitive with a large ^{3}He based system. Moreover, the realization of large, cost effective neutron detectors could, for the first time, enable the detection of multiple neutrons per fission from within a large object such as a cargo container. Such a signal would provide a robust indication of the presence of fissioning material, reducing the frequency of false alarms while increasing sensitivity.

Keywords: Water Cerenkov, neutron detector, gadolinium, neutron capture

1. Introduction:

The detection of a significant quantity of Special Nuclear Material (SNM), such as plutonium or uranium, heavily shielded inside a cargo or other container presents a unique challenge. Gamma-ray emissions from such sources tend to be low in energy and/or flux. Gamma rays are also relatively easy to shield with high Z material. Shielding neutron emission is a more difficult proposition; nevertheless, neutrons can be moderated to the extent that directional and spectral information is lost. Only a few technologies can meet the very specific requirements imposed by the physics. One key approach is to maximize solid angle coverage: for example, it may be possible to look for coincident neutron emission with a detector that almost completely surrounds the object of interest. With the technologies currently available, this would present an expensive and complicated task: ^{3}He is in short supply, BF_3 tubes and liquid scintillator are highly toxic and/or flammable and ^{10}B tubes have relatively low efficiency.

* Corresponding Author. Tel: +1 925 423 4792. Email: dazeley2@llnl.gov

The Advanced Detectors Group at LLNL has been developing a cost effective technology that may serve as a large solid angle detector for portal monitors or other applications. We use pure water doped with gadolinium tri-chloride ($GdCl_3$), which serves both as a moderator for neutrons and a detection medium. Moderated neutrons that capture on a gadolinium nucleus produce a shower of gamma rays, whose energies add to a total of approximately 8 MeV. Subsequent Compton scattering and Cherenkov light emission resulting from these gamma rays is detected by an array of large PMTs.

In 2010 Kouzes *et al*. [3] described in detail the three minimum characteristics required of a real world RPM neutron detector. The requirements are: 1) better than 2.5 cps/ng of ^{252}Cf absolute efficiency (source 2m away), 2) intrinsic ^{60}Co gamma-ray discrimination better than 10^{-6}, and 3) consistent neutron detection efficiency under conditions of high ambient gamma-ray exposure of up to 10 mR/h. The neutron detection technologies available until now tend to be expensive and do not allow for sufficient solid angle coverage for correlated detections, particularly from large objects such as cargo containers. Perhaps for this reason, the above list of characteristics does not include an explicit standard for sensitivity to time-correlated neutrons, which would in turn require large detectors. However, time-correlated neutron multiples are a highly unusual signature of SNM, and their efficient detection would provide significant additional background rejection capability relative to current RPMs, which rely only on an overall enhanced rate of neutrons or gamma-rays as a detection strategy. Detection of time-correlated neutrons – arising from single fission events or from fission chains in weakly multiplying SNM - is possible if a detector of sufficient solid angle and efficiency can be developed at a reasonable cost. In this paper we present a design that could enable a new generation of large, cost effective, highly efficient, low background neutron detectors suitable for RPM and similar applications.

Our group has built and successfully operated two tonne-scale neutron detectors. A 3.5 kiloliter detector was completed in 2009 [2] and a 1-kiloliter antineutrino detector the following year. We have developed a Geant4 simulation that reproduces the performance of both detectors; a detailed description of the simulation will be given in Section 2. The larger of the two detectors had poorer than anticipated water quality caused by UV stabilizing agents leaching into the water from the polyethylene tank. Many manufacturers include UV stabilizers in their polyethylene to reduce long-term sun damage. UV stabilizers do indeed leach into DI water very quickly as shown by [4]. This, when combined with a larger volume, reduced the energy resolution. Despite this, the detector achieved neutron detection efficiencies of 25% to 30% before analysis cuts were applied. The 1-kiloliter detector incorporated some important improvements compared to the 3.5- kiloliter detector. The most important was the use of a stainless steel tank, rather than polyethylene. The chemical compatibility of $GdCl_3$ and stainless steel, identified as a problem in the past [5], was mitigated by coating the interior wall with baked-on Teflon. Another improvement was the installation of a highly reflective layer of Teflon, GORE-DRP, around the walls of the tank. The manufacturer claims that GORE-DRP can achieve a diffuse reflectivity of better than 99%. Stainless steel construction was an advantage in terms of detector robustness and mobility.

These improvements had an immediate impact on both the water quality and detector energy resolution. Figure 1 shows a comparison of detector response spectra from neutron captures inside both the 3.5-kiloliter detector (a) and the 1-kiloliter detector (b) using a ^{252}Cf fission source. In the presence of a fission neutron source, whenever there

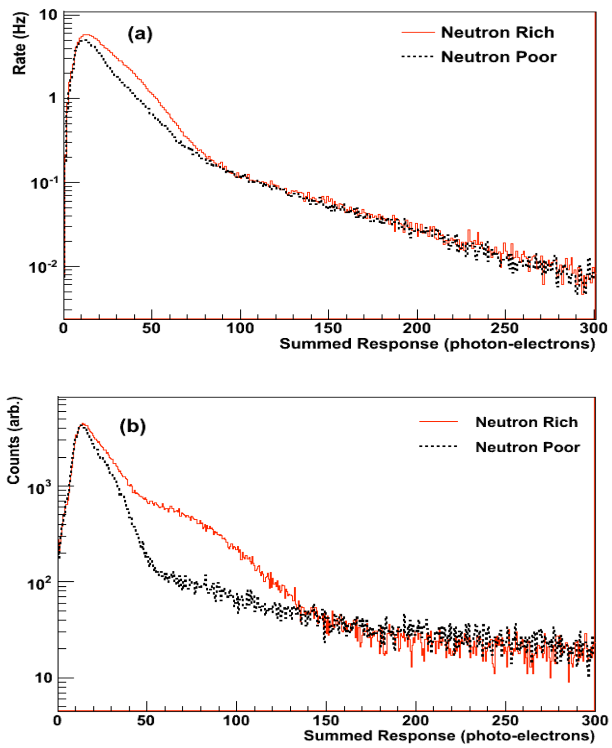

Figure 1: The detector response, in terms of detected photoelectrons, of both the 3.5 kiloliter (a) and 1 kiloliter neutron detectors (b). The top figure, (a) was taken from [2].

are two events correlated in time, the second event is more likely to be that of a neutron capture. We show a comparison of the detector response due to these correlated events (red curve), together with a similar selection of uncorrelated events (black curve). The red and black curves are normalized with equal numbers of uncorrelated (background) events. All the spectra have the same generalized features: a peak at low energies due primarily to low energy gamma rays, commonly referred to as Naturally Occurring Radioactive Material (NORM), incident on the tanks from the surrounding environment, and a high energy tail due to muons and cosmogenic gamma-rays. An important consideration for any neutron detector technology is that low energy gamma rays do not interfere with the neutron detector signal: the separation between neutrons and gamma rays observed in the 1-kiloliter detector bodes very well for this technology.

2. The Simulation

We have developed a Geant4 simulation to model the response of both tonne-scale detectors. The main differences between the two models are the water quality and the wall reflectivity, both of which were significantly lower in the 3.5-kiloliter detector. The underlying physics and PMT responses are the same in both simulations. Figure 2 shows a schematic of both detectors. As discussed above, the water quality in the large detector had a number of issues. With respect to simulation tuning, having two different detectors with different optical qualities was actually an advantage.

In Figure 3, we present a comparison of the simulated and measured detector response to neutron capture from a source mounted flush against the side wall of the 3.5-kiloliter detector. Neutron captures resulting from such a configuration tend to capture near the wall of the detector, resulting in a partial loss of capture gamma rays from the detector, reducing the overall response relative to neutron captures in the center. Figure 4 shows a similar comparison of neutron captures throughout the 1-kiloliter detector volume.

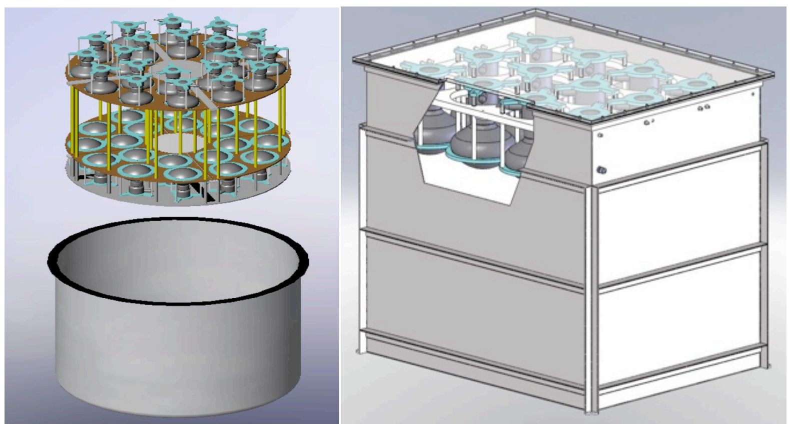

Figure 2: Schematic designs of both the 3.5 kiloliter (left) and 1 kiloliter (right) water based neutron detectors.

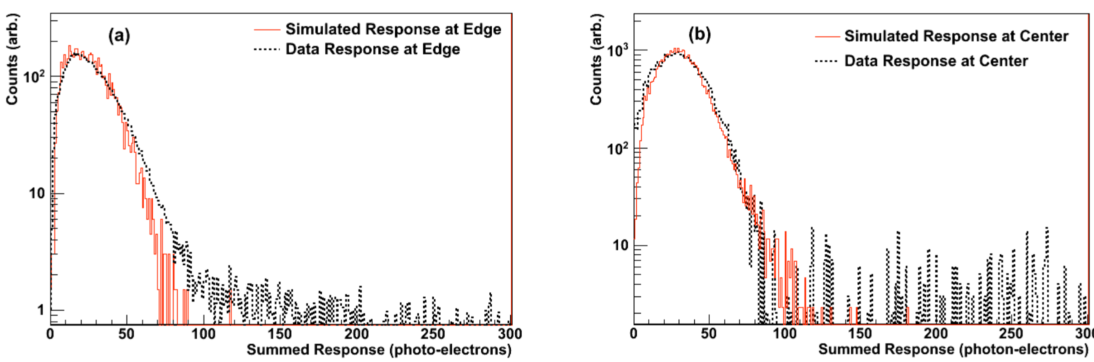

Figure 3: A comparison of Monte Carlo based detector response to neutron captures (red) in the 3.5-kiloliter detector and real data (black). In the real data, the detector response to neutron captures is revealed by applying a statistical subtraction of uncorrelated events from a selection of neutron rich correlated events (see Figure 1). Taken from [2].

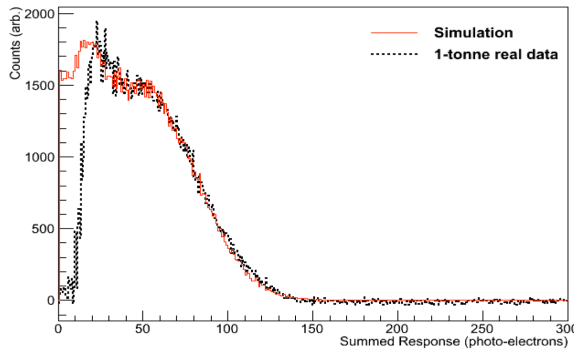

Figure 4: Comparison of real and simulated neutron captures throughout the volume of the 1-kiloliter detector. The real data spectrum is a statistical subtraction of the two curves in Figure 1(b): it cuts off at low energies as a result of the trigger threshold, which is not modeled in the simulation.

2.1 Wavelength Shifter

The primary issue with Cherenkov detectors is the low light output, and hence low energy resolution relative to liquid organic scintillator. We recently reported some progress in this area [6], studying the relative gain in light detected from cosmic muons propagating downwards through a 250 liter acrylic tank instrumented with 6 eight inch PMTs and filled with water and doped with three different water soluble wavelength shifters. The detected Cherenkov signal was approximately doubled with the addition of 1 part per million of wavelength shifter 4-Methylumbelliferone (4-MU). The observed gain also proved stable over a period of a few months. The gain dependence on wavelength shifter concentration is shown in Figure 5 together with our simulation. Qualitatively we see that the simulation agrees with the experiment - the gain begins to saturate at approximately a factor of two at 1ppm. The other two chemicals we tested were Carbostyril-124 and Amino-G acid, neither of which were the equal to 4-MU in terms of long term stability, dissolvability and gain. Having settled on 1ppm 4-MU as our wavelength shifter of choice, we tested it in the 3.5-kiloliter detector. Figure 5 shows

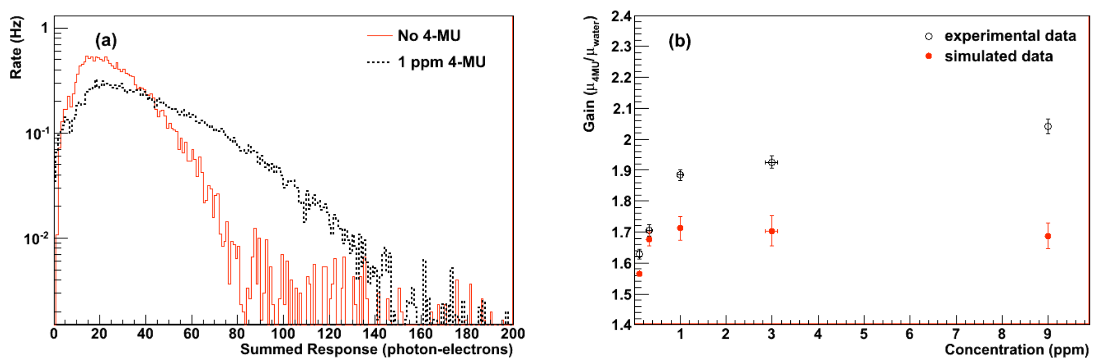

Figure 5: (a) The detected neutron capture spectrum before and after the addition of 1ppm of 4-MU in the 3.5-kiloliter detector: the spectrum is plotted in terms of detected photoelectrons. The relative gain in detected light due to 1ppm of 4-MU wavelength shifter is presented in (b). From [6].

that we again observed a factor of approximately 2x improvement in gain, this time in the neutron capture spectrum. This was again replicated in both the simulation and real data.

3. Optimized Water-based Neutron Detector System

The following basic design principles incorporate many of the lessons learned from our earlier neutron detector systems and incorporate additional optimizations suited to a field deployable RPM. These are:

I. Optimum size and portability – small enough to be moved by hand with a pallet jack, large enough to contain a sufficient fraction of gamma-rays resulting from neutron capture (~0.5 to 1 tonne modules)
II. Tank materials ALL Teflon/polypropylene – We have great deal of evidence now that these two materials are compatible with gadolinium-doped water over time periods of a year or so
III. High gadolinium concentration – shortened inter-event times between correlated pairs of neutrons, reducing uncorrelated gamma-ray NORM background
IV. Segmented volume – a small gadolinium doped section covering one full face of each module and oriented towards the potential SNM source will reduce the number of detected cosmogenic and other background neutrons). The detection of groups of neutrons initiated via spallation within the detector volume itself will be reduced in number by a factor proportional to the relative volumes of gadolinium and pure water. The detection of neutrons initiated outside the detector would be reduced by a greater factor
V. Wavelength shifters – increased energy resolution, increasing efficiency while reducing backgrounds
VI. Muon veto – reduced cosmogenic sources of neutrons and eliminates a class of background caused by muons that clip the edge of the detector depositing an equivalent amount of energy as a neutron.
VII. High solid angle coverage – ideal for correlated neutron search and a well shielded source

Figure 6 shows a schematic of a single cubic meter module duplicated in our simulation. We show 9 PMTs mounted at the top, a 0.6% $GdCl_3$ doped segment situated at the front of the detector comprising about 1/5th of its volume and physically contained within UV transmitting acrylic. A plastic scintillator muon veto system would fully surround the detector. A system of modules could in principle include as many as needed, depending on the desired solid angle coverage. Employing multiple separate modules and requiring the presence of correlated events in different modules may also be an effective approach for reducing cosmogenic neutrons. The Geant4 simulation presented here and elsewhere ([2],[6]) comprises a system of 4 such modules situated on either side of a 3-meter wide lane of traffic to gauge its likely performance.

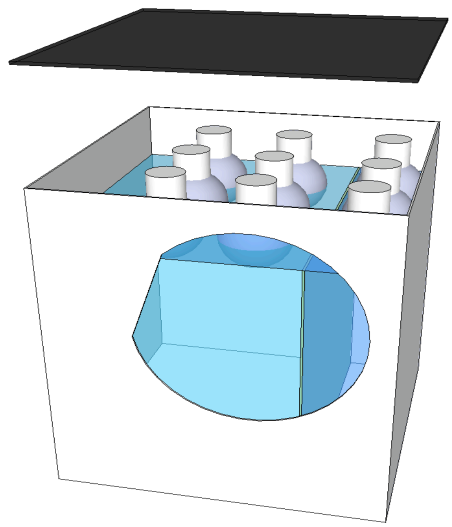

Figure 6: A schematic showing the design of the 1-tonne water neutron detector modules of our simulation. The module contains 9 PMTs and two distinct volumes separated by an acrylic wall. On the right hand side the small volume is neutron sensitive and contains pure DI water doped with 0.6% $GdCl_3$. The left hand side is larger and contains pure DI water. A muon veto system surrounds this detector (not shown)

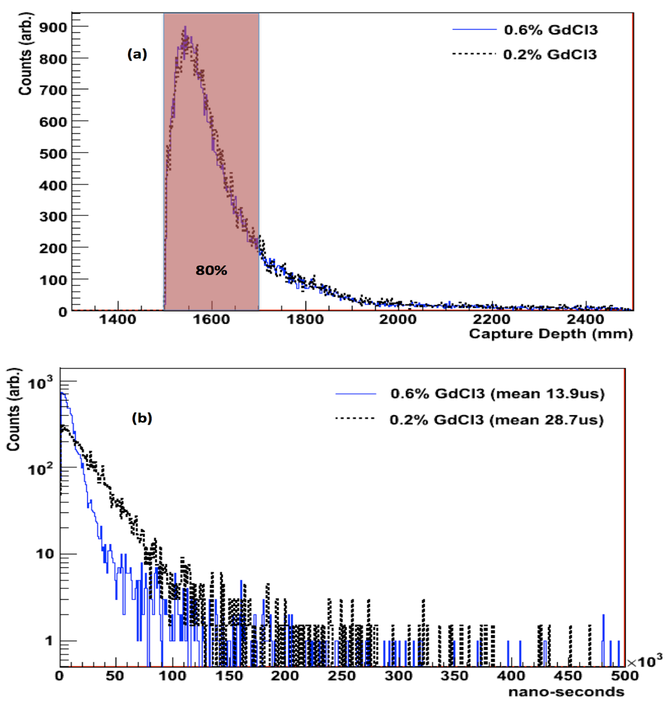

Figure 7: The neutron capture depths within each module for a fission source location central to the four module system (a), and the inter-event time distributions (b) for different concentrations of $GdCl_3$ obtained from the simulation. The mean inter-event times were 14 microseconds and 29 microseconds for 0.6% and 0.2% $GdCl_3$ respectively. The red shaded region in (a) indicates a depth of 20 cm from the detector edge.

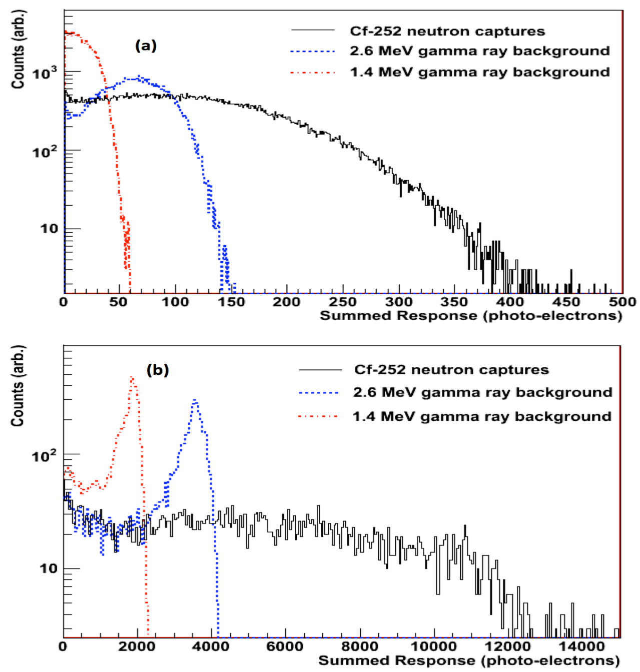

Figure 8: Detector response to neutron captures, 2.6 MeV gamma ray backgrounds and 1.4 MeV gamma ray backgrounds in both an optimized water Cherenkov detector (a) and a similar detector filled with liquid organic scintillator plus gadolinium (b). Note that the energy resolution of neutron capture events in both cases is very similar (and poor), which implies that only a marginal improvement in detector efficiency can be achieved by employing liquid scintillator.

Figure 7 shows the neutron capture locations in terms of detector depth (a) and the inter-event time distribution (b) obtained from 0.6% and 0.2% $GdCl_3$. Figure 7(a) indicates approximately 80% of the neutrons from a centrally located source would be captured within the 20 cm deep gadolinium doped detector segment. This was not dependent on the gadolinium concentration. The use of 0.6% $GdCl_3$ reduces the mean inter-event time from 29 to 14 microseconds (see Figure 7(b)). As pointed out by [7], the relative capture times of two correlated fast neutrons follow an exponential distribution, as they both must thermalize before one captures. Given that, the $GdCl_3$ level will be limited only by water quality.

If a 50% reduction in capture time is achievable, it could have a large impact on the correlated event background, as it would approximately halve any inter-event time cuts employed in analysis, proportionately reducing the prevalence of random correlations caused by high-energy cosmogenic gamma rays or muons. In fact, this single factor constitutes our main tool to reduce high-energy cosmogenic gamma rays, which cannot be confronted via a muon veto. In our earlier study of $GdCl_3$ doped water attenuation, we found no evidence of any degradation at a concentration of 0.2%. We think it is therefore unlikely that a concentration of approximately three times that will have a large impact on detectors at the cubic meter scale.

Using our simulation, adjusted to model the 4 x optimized module configuration (two modules arranged on both sides of a lane of traffic), we estimated its likely performance. We assumed a relatively conservative water attenuation length of 25 meters (Pure DI water is approximately 100 meters [8]). Figure 8 shows the total detector response of each module (sum of all 9 PMT signals), obtained assuming an isotropic background of 1.4 and 2.6 MeV gamma-rays and a flux of ~1 MeV neutrons from a centrally located fission source.

Table 1 shows the predicted signal and background of the simulated 4-module system assuming that it collects data for 20 seconds per cargo container. The signal assumes a 3 micro-Curie ^{252}Cf fission neutron source. The signal event rates were obtained from the simulation, and the background rates were extrapolated from our single 1-kiloliter detector measurements (1500Hz event rate per module). The charge analysis cuts (150 to 400 photoelectrons) were chosen to eliminate (nearly) all low energy terrestrial gamma rays of 2.6 MeV or less. The physics events capable of passing these analysis cuts are dominated by genuine neutron captures, muons or higher energy cosmogenic gamma rays. We assume for simplicity that the muon veto efficiency is 100%. The high-energy cosmogenic gamma ray flux at sea level is ~100 $m^{-2}s^{-1}$ depending on the low energy cutoff (e.g. [9]). The background rates in Table 1 were scaled from our 1-kiloliter detector measurements, which employed a charge cut of between 50 and 150 photoelectrons to remove the low energy gamma ray background, obtaining a rate of 130Hz, consistent with the above prediction of ~100Hz m^{-2}, and 8.7% of our measured flux. At these rates, 30k background events pass our charge analysis cut in a 20 second detector dwell time. Approximately 2/3 of those are due to muons. Note that uncorrelated neutron detector systems can be susceptible to variations in background over time. Our system, which measures variations in both the uncorrelated and correlated neutron rate, will produce a more robust signal from a hidden fission source.

Table 1: The number of events detected in our simulation of the 4-module system of cubic meter detectors in a dwell time of 20 seconds. The neutron source simulated was a 3 micro-Curie ^{252}Cf fission source producing a mean of 3.75 neutrons per fission (producing approximately 200,000 neutrons in total). Operating in correlated mode the signal detected (101 correlated event pairs) represents a 5.7σ excess over background (107 event pairs).

Background Reduction Method	**3 micro-Curie ^{252}Cf Emission 200 000 neutrons**		**Non-Neutron Background**	
	# singles events	# event pairs	# singles events	# event pairs
None	15.6k	2.69k	120k	120k
150pe<charge<400pe	4.02k	187	30k	30k
dT<20 micro-seconds	N/A	168	N/A	894
20 cm gadiated region (80.4% efficiency) *	3.23k	108	30k	894
Muon Veto (6% dead time)	3.04k	101	9.8k	107

*** Detector segmentation into gadolinium and non-gadolinium doped regions reduces neutron backgrounds only**

Detector Energy Resolution

The primary factor influencing the resolution of this detector is not, as one would expect, the low light output from the water Cherenkov mechanism, but is the escape of neutron capture gamma rays from the detector volume. This is illustrated in Figure 8. We simulated neutron capture, 2.6 MeV and 1.4 MeV gamma rays within both a cubic meter water based module and a hypothetical cubic meter of gadolinium doped liquid organic scintillator. In both cases the gadolinium capture peak appears as a wide unresolved peak that extends over the full range of detector response from 0 to 8 MeV. The gamma-ray peaks of course *were* resolved very well in the scintillator system. However, the relative increase in efficiency that one might achieve after applying an energy cut to remove these backgrounds is not significantly higher in the scintillator system than the water system. The scintillator-based system would come at the cost of a massive reduction in detector operability in high background environments due to pile up.

Conclusion:

Water based neutron detectors offer the potential for scalable, highly efficient, and cost effective radiation portal monitors. We have built and fully characterized two such detectors and built a Geant4 based model described here (and [2],[6]) that successfully reproduces their performance characteristics. Using this model as a baseline, we have incorporated a number of optimizations and efficiencies that have separately been effective in improving detector response, but have heretofore not been integrated into a single system. The simulation predicts an optimized 1-tonne scale module would be capable of 25% fast neutron detection efficiency with negligible susceptibility to low energy background gamma-rays (< ~1MeV). With this performance the minimum detectable amount of plutonium (93% ^{239}Pu, 7% ^{240}Pu) our 4-module system is capable of detecting is 75 grams (to 3σ), within a 20 second dwell time. A significant signal would be detectable in both correlated and uncorrelated mode. Additionally, since this system is sensitive to correlated neutrons, its performance would scale with the square of the solid angle coverage. Based on this work, in the near future we plan to build a 4-module system to test in a number of real world scenarios. Testing will include the presence of neutron and gamma-ray shielding, large metal objects such as a truck, and finally the presence of high background, low energy NORM type radiation. Detecting well-shielded SNM within a cargo container remains a challenge for present day technology, particularly with the recent shortage of ^{3}He. The system described here may represent a low cost, reliable, environmentally benign, and efficient solution.

Acknowledgements:

The authors would also like to thank John Steele for programming the trigger logic in the FPGA and Dennis Carr for valuable help during the design and construction of both of our neutron detectors. This work was performed under the auspices of the US Department of Energy by Lawrence Livermore National Laboratory under contract DE-AC52-07NA27344, release number LLNL-xxxxxx. The authors wish to thank the DOE NA-22 for their support of this project.

References:

[1] “Observation of neutrons with a Gadolinium doped water Cherenkov detector”, S. Dazeley, A. Bernstein, N. S. Bowden, R. Svoboda., Nucl. Inst. And Meth. A, V607, pp616 (2009)

[2] “Large-scale gadolinium-doped water Cherenkov detector for nonproliferation”, M. Sweany *et al*., Nucl. Inst. And Meth. A. V654, pp.377, (2011)

[3] “Neutron detection alternatives to ^{3}He for national security applications”, R. T. Kouzes *et al*., Nucl. Inst. And Meth. A, V623, P1035 (2010)

[4] “Measurement of the Absolute Attenuation Length of UV Light in Pure and Material-Soaked Water Using an Horizontal Photometer”, S. Ouedraogo *et al*., Submitted to Nucl. Instr. and Meth. A, (2012).

[5] “Transparency of 0.2% $GdCl_3$ doped water in a stainless steel test environment”, W. Coleman, A. Bernstein, S. Dazeley and R. Svoboda, Nucl. Instr. and Meth. A, vol. 595 pp. 339-345, (2008)

[6] “Study of wavelength-shifting chemicals for use in large-scale water Cherenkov detectors”, M. Sweany *et al*. Nucl. Instr. and Meth. A, vol. 664, pp 245-250, (2012)

[7] “A Note on Neutron Capture Correlation Signals, Backgrounds, and Efficiencies”, N. S. Bowden, M. Sweany, S. Dazeley, Submitted to Nucl. Instr. and Meth. A, (2012).

[8] “Absorption spectrum (340 -640 nm) of pure water. I. Photothermal measurements”, F. M. Sogandares and E. S. Fry Applied Optics vol. 36, pp. 8699 (1997)

[9] “The Atmospheric Gamma-Ray Spectrum From 50 MeV to 1 GeV at 3 mbar and Sea Level”, J. Staib, G. Frye, A. Zych, Proceedings of the 13th International Conference on Cosmic Rays, Denver, Vol 2, pp 916, (1973)